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The requested laser system was purchased. All of the funds ($155,782) were used for the purchase of a Coherent 900-F tunable Ti:sapphire laser system with a Verdi 10 pump laser, in conjunction with matching funds that came from Boston College. The total cost of this laser system was $191,500. The laser system was installed successfully during the project period and is currently being used to study how the efficiency of data storage and retrieval in our media depends on the wavelength of light employed.

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Final Performance Report

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Award F49620-02-1-0273

"Request for Funds for the Purchase of a Broadly-Tunable Ti:sapphire Laser for Research in High-Density Optical Memory"

Objectives: The objective of this project was to acquire a broadly-tunable Ti:sapphire laser system and associated optics for the spectroscopic study of optical data storage in molecular glasses and highly-crosslinked polymer. The purpose of the broadly-tunable laser system is to be able to vastly increase our capability for characterizing the data storage materials. While it is possible to create a large amount of fluorescent product either thermally or with ultraviolet light, the photoproducts that are responsible for data storage appear to be essentially unique to the mulitphoton excitation process, or at least are produced in such small amounts by the other processes that it is impossible to analyze them. On the other hand, it is not possible to create large amounts of photoproduct in a process that relies on the use of sub-micron focal spots. Thus, the only truly viable technique for analyzing the products is performing in-situ spectroscopy on the product in femtoliter volumes. This requires a highly-sensitive detection system, which we have, as well as the ability to tune the writing and readout laser sources; it is the latter capability that was provided by the laser system purchased under this grant. The idea is to be able to map out the three-photon excitation spectrum of the storage materials and to map out the two-photon excitation spectrum of the photoproduct. This will not only assist in the spectroscopic determination of the identity of the product species, but will also allow us to optimize the storage and readout processes. For instance, we can find the optimum wavelength for rapid data storage with three-photon excitation, or look for a wavelength at which readout can be accomplished with two photons whereas three photons do not provide sufficient energy to store data. With the new laser system in combination with the existing one in our laboratory, the point was to have the capability to write data at one wavelength and then to read them immediately at another.

Status of Effort: The requested laser system was purchased. All of the funds ($155,782) were used for the purchase of a Coherent 900-F tunable Ti:sapphire laser system with a Verdi 10 pump laser. Matching funds from Boston College allowed us to purchase steering optics, Faraday isolators, and other equipment so that we can use both the new broadly-tunable laser and our older, non-tunable laser simultaneously in the multiphoton microscope. This allows us to write data at one wavelength and to read them out immediately at another wavelength, for instance. The total cost of this laser system was $191,500. The laser system was installed successfully during the project period. A photograph of the completely installed laser system with the cover off is shown in Figure 1.

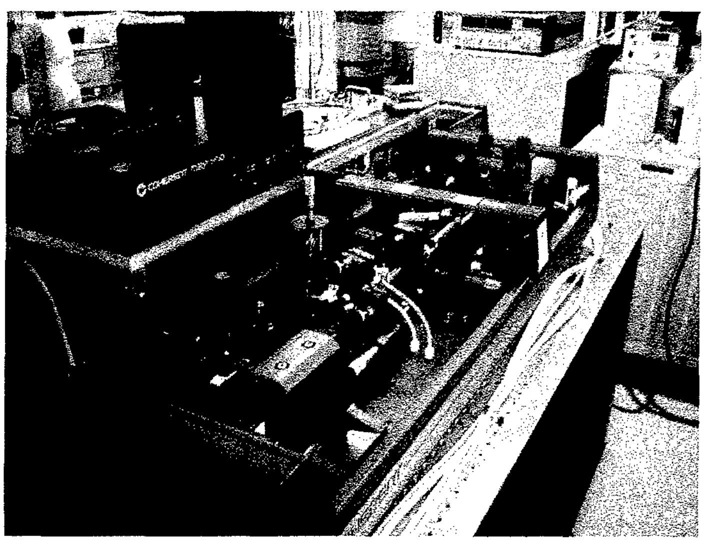

Figure 1. A photograph of the Mira 900-F. The Verdi-10 pump laser is inside the box in the upper middle portion of the image.

The laser system offers ready tunability from 700 nm to 900 nm, as shown in Figure 2. With nitrogen purging of the laser cavity the tunability with the main mirror set extends out to about 980 nm, and with a different mirror set in combination with purging the wavelength range extends out to 1.1 microns.

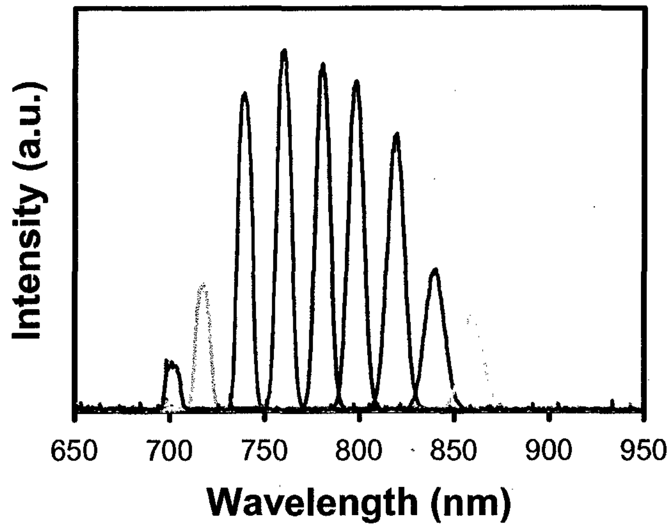

Figure 2. Representative laser spectra over much of the tuning curve of the laser with the broadband mirror set and an unpurged cavity.

Initial experiments in testing the dependence of the readout and storage processes on wavelength proved not to be highly reproducible. The difficulty proved to be that tuning the laser could alter the pulse duration substantially. Since the three-photon storage process depends on the intensity cubed and the two-photon readout process depends on the intensity squared, both are highly sensitive to changes in pulse duration (and therefore in peak intensity). Thus, performing the experiments reproducibly requires that we be able to exert good control not over the intensity, but also the duration of the laser pulses. The microscope optics can be highly dispersive, and so it is crucial that these properties be measured *at the sample position,* which is not a simple problem. We took our cue for doing this from previous work that demonstrated that two-photon photoconductivity in gallium arsenide photodiodes could be used to accomplish this task. However, previous work relied on performing interferometric autocorrelations, which require stepping in increments that are considerably longer than the wavelength of the light used. This is a slow and noisy process for laser pulses that are not a few tens of femtoseconds long, and makes adjustment of the pulse length highly laborious. We have instead designed an optical system that allows for the measurement of laser pulses with parallel but non-

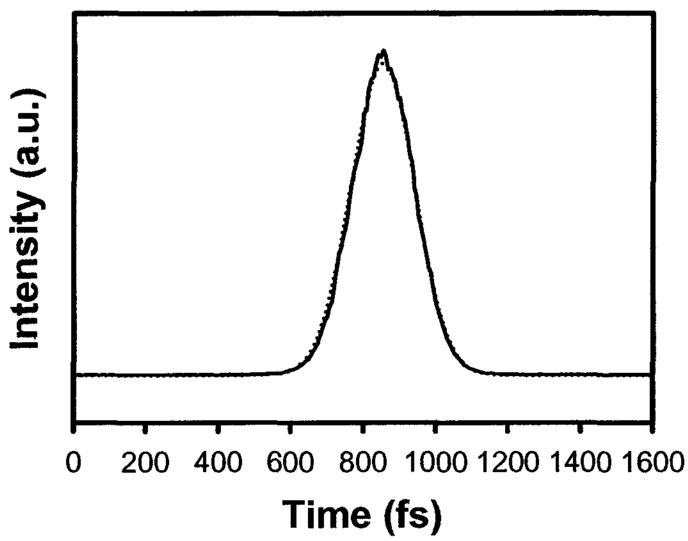

Figure 3. Representative non-interferometric auto-correlation (solid line) and gaussian fit (dotted line). The pulse width in this case is 146 fs.

collinear beams. In this case the autocorrelation is not interferometric, and much larger step sizes can be taken. Because interferometric stability is not required, the autocorrelations are much less noisy as well. The combination of these two advantages allows us to obtain autocorrelations rapidly with high signal-to-noise ratios (Figure 3), making it simple to adjust the dispersion compensation of the laser system to adjust the pulse length to the desired value. This autocorrelation system is the subject of a manuscript that will soon be submitted.

Given these advances, we are now to the point of being able to collect reliable and reproducible data on the wavelength dependence of data storage and retrieval in our molecular glasses and highly crosslinked polymers, and these experiments are in progress. The equipment purchased with this grant has been and will continue to be of tremendous importance to this project.